

Extension of the Lieb-Schupp theorem to the Heisenberg models with higher order interactions

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We extend the Lieb-Schupp theorem to the Heisenberg models with higher order interactions on non-frustrated or frustrated finite lattices. These lattices are constructed by even numbered rings with or without crossing bonds and have reflection symmetry. The results show that all ground states have total spin zero in wide interaction parameters region which is not covered with the results of the Marshall-Lieb-Mattis type arguments.

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I. INTRODUCTION

The Heisenberg models with higher order interactions have been discussed from various points of view. Hamiltonian of the simplest model consists of bilinear and biquadratic exchange interaction terms. The $S = 1$ bilinear-biquadratic exchange interaction model has been investigated in the context of the Haldane conjecture[1] and as models of one dimensional spin-Peierls material $\text{Li}_2\text{VGe}_2\text{O}_6$, [2, 3] two dimensional gapless spin liquid material NiGa_2S_4 , [4–6] magnetism in $S = 1$ bosons [7, 8] and three-flavor fermions [9] trapped in optical lattices, magnetism in iron pnictide superconductors [10, 11], and deconfined criticality and Landau forbidden phase transition [12–14]. In the case of $S = 3/2$ this model is also investigated as a model of the chromium spinel oxides ACr_2O_4 ($A = \text{Hg}, \text{Cd}, \text{Zn}$). [15, 16] The $S = 3/2$ bilinear-biquadratic-bicubic exchange interaction model is studied as a model of magnetism in $S = 3/2$ fermions trapped in optical lattices [17–19] and as a resource of the measurement based quantum computer. [20, 21] In particular the $S = 1$ model was extensively studied by theoretical works, but models with $S \geq 3/2$ and/or bicubic and more higher order interactions are less studied and is of importance to the understanding of magnetic properties of the chromium spinel oxides and cold atomic gases in optical lattices.

The Marshall-Lieb-Mattis theorem is one of the most famous exact results of quantum spin systems. In the case of the antiferromagnetic Heisenberg model on bipartite lattices with the same number of sublattice points, it proved that ordering energy levels, i.e., the lowest energy level for allowed total spin is monotonically increasing function of total spin and ground state is unique spin singlet. [22] This theorem was extended to the case of the spin- S bilinear-biquadratic exchange interaction model. [23–25]

The Marshall-Lieb-Mattis theorem has made a lasting contribution to check validity of a huge number of

results for the numerical studies of quantum spin systems on bipartite lattices by using the exact diagonalization, density matrix renormalization group, quantum Monte Carlo simulation, etc., and now it serves as guidelines for ‘*real quantum simulators*’ envisioned by Richard Feynman. [26, 27] But this theorem is not applicable to the models on non-bipartite or frustrated lattices. In 1999 Lieb-Schupp succeeded to prove that ground states of the antiferromagnetic Heisenberg model on checkerboard type of the square lattice with crossing bonds have total spin zero. [28–30] Their method use reflection symmetry of Hamiltonian, on the other hand, the Marshall-Lieb-Mattis theorem is based on the Perron-Frobenius theorem and works well if it can be find suitable unitary transformation which leads to same sign of off-diagonal matrix elements of irreducible unitarily transformed Hamiltonian satisfying the Perron-Frobenius theorem. But it seems that there is no systematic method available to find it so far. The Lieb-Schupp theorem can be applied to a class of frustrated spin systems on reflection symmetric lattices, but it can not give any information for the degeneracy of the ground state.

Our purpose in the present paper is an extension of the Lieb-Schupp theorem to the Heisenberg models with higher order interactions on finite size lattices which are constructed by even numbered rings with or without crossing bonds and have reflection symmetry. As explained above, the Marshall-Lieb-Mattis type argument does not work for non-bipartite lattices. Adding antiferromagnetic crossing bonds to even numbered rings induces a frustration of Néel order and breaks bipartiteness of lattices, but their reflection symmetry are preserved. By using this nature of lattices we will prove that all ground states of these models possess total spin zero in wide parameter region which is not covered with results of the Marshall-Lieb-Mattis type arguments.

This paper is organized as follows. In section II, we introduce some definitions and notation used throughout this paper. In section III, to keep the paper self-contained, we explain a basic setup and ideas of the Lieb-Schupp theorem and apply this theorem to the models on even numbered rings to prove that all ground states have

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total spin zero. In section IV, with Hamiltonian on even numbered rings discussed in section III as a local Hamiltonian, we construct global Hamiltonian on two dimensional lattices. In particular we treat square and honeycomb lattices with crossing bonds. In section V, we summarize and discuss the results of sections III and IV and comment on the effects of the crossing bonds in infinite system of the $S = 1$ bilinear-biquadratic exchange interaction model and physical realization of ferroquadrupole (spin nematic) phase in magnetic materials.

II. DEFINITION AND NOTATION

In this paper we study the isotropic spin- S Heisenberg model with up to the n -th order ($1 \leq n \ll \infty$) interaction term:

$$H_n = - \sum_{x \neq y \in \Lambda} \sum_{k=1}^n J_k(|x-y|) (\mathbf{S}(x) \cdot \mathbf{S}(y))^k, \quad (1)$$

on lattice Λ , where $-J_k(|x-y|)$ are the k -th order interaction coefficients between sites x and $y \in \Lambda$. The summation over $x \neq y \in \Lambda$ counts every pair (once and once only). $\mathbf{S}(x) = (S_1(x), S_2(x), S_3(x))$ denotes spin- S operator on site x and satisfies the usual commutation relations:

$$[S_i(x), S_j(y)] = i\epsilon_{ijk} S_k(x) \delta_{xy}. \quad (2)$$

Here we use a usual basis in which $S_3(x)$ is diagonalized, $S_1(x)$ and $S_3(x)$ have real matrix elements and $S_2(x)$ pure imaginary.

This Hamiltonian can be written as the spin- S isotropic Hamiltonian with up to 2^n -pole interaction term:

$$\mathcal{H}_n = - \sum_{x \neq y \in \Lambda} \sum_{k=1}^n I_k(|x-y|) \sum_{q=-k}^k O_{k,q}(x) O_{k,q}^\dagger(y), \quad (3)$$

where the Racah operators $O_{k,q}(x)$ (2^k -pole operators) satisfy the relations:

$$O_{k,k}(x) = \frac{(-1)^k}{2^k k!} [(2k)!]^{1/2} (S_+(x))^k, \quad (4)$$

$$O_{k,q}^\dagger(x) = (-1)^q O_{k,-q}(x), \quad (5)$$

$$[S_3(x), O_{k,q}(y)] = q O_{k,q}(x) \delta_{xy}, \quad (6)$$

$$[S_\pm(x), O_{k,q}(y)] = \sqrt{k(k+1) - q(q \pm 1)} O_{k,q \pm 1}(x) \delta_{xy}, \quad (7)$$

with $S_\pm(x) = S_1(x) \pm iS_2(x)$ and $-k \leq q \leq k$. $-I_k(|x-y|)$ are the 2^k -pole interaction coefficients between sites x and y . Relations between multipole interactions and higher powers of Heisenberg interaction are

known to be

$$\sum_{q=-1}^1 O_{1,q}(x) O_{1,q}^\dagger(y) = \mathbf{S}(x) \cdot \mathbf{S}(y), \quad (8)$$

$$\begin{aligned} & \sum_{q=-2}^2 O_{2,q}(x) O_{2,q}^\dagger(y) \\ &= \frac{3}{2} (\mathbf{S}(x) \cdot \mathbf{S}(y))^2 + \frac{3}{4} \mathbf{S}(x) \cdot \mathbf{S}(y) - \frac{1}{2} S^2 (S+1)^2, \end{aligned} \quad (9)$$

and for $k \geq 3$ they are given by equations (B.20) and (B.21) in reference [32]. So H_2 is written as

$$\begin{aligned} H_2 = & - \sum_{x \neq y \in \Lambda} I_1(|x-y|) \sum_{q=-1}^1 O_{1,q}(x) O_{1,q}^\dagger(y) \\ & - \sum_{x \neq y \in \Lambda} I_2(|x-y|) \sum_{q=-2}^2 O_{2,q}(x) O_{2,q}^\dagger(y) \end{aligned} \quad (10)$$

with

$$I_1(|x-y|) = J_1(|x-y|) - \frac{J_2(|x-y|)}{2}, \quad (11)$$

$$I_2(|x-y|) = \frac{2}{3} J_2(|x-y|), \quad (12)$$

where we have omitted a constant term in the right hand side of equation (10).

Let us also introduce the total spin operator:

$$\begin{aligned} \mathbf{S}^{\text{tot}} &= (S_1^{\text{tot}}, S_2^{\text{tot}}, S_3^{\text{tot}}) \\ &= \left(\sum_{x \in \Lambda} S_1(x), \sum_{x \in \Lambda} S_2(x), \sum_{x \in \Lambda} S_3(x) \right). \end{aligned} \quad (13)$$

We easily see continuous symmetry of \mathcal{H}_n :

$$[S_1^{\text{tot}}, \mathcal{H}_n] = [S_2^{\text{tot}}, \mathcal{H}_n] = [S_3^{\text{tot}}, \mathcal{H}_n] = 0. \quad (14)$$

III. MODELS ON EVEN NUMBERED RINGS

In this section we discuss conditions for establishment of the Lieb-Schupp theorem which applies to Hamiltonian (3) on even numbered rings and properties of its ground state.

Before we move forward, let us explain a setup of finite size lattices Λ which is needed to establish the Lieb-Schupp theorem. Throughout this paper we consider $\Lambda = \Lambda_L \cup \Lambda_R$ which has an even number of independent sites and Λ can be split in two equal parts Λ_L and Λ_R . Λ_L and Λ_R are mirror images of one another about a symmetry plane without sites which cuts bonds between sites $x \in \Lambda_L$ and $y \in \Lambda_R$, and the collection of these sites is denoted by Λ_C if Λ is single even numbered rings.

In the following we treat the models on just single even numbered rings, i.e., closed chains with even number of sites, to prepare constructions of the models on two dimensional lattices in section IV.

A. Ground state of models on even numbered rings and their reflection symmetry

Following the above manner let us write Hamiltonian (3) on single even numbered rings with $2m$ sites:

$$h_{\text{ring}}^{2m} = h_L^m + h_R^m + h_C^{2m}, \quad (15)$$

where

$$h_L^m = - \sum_{x \neq y \in \Lambda_L} \sum_{k=1}^n \sum_{q=-k}^k I_k(|x-y|) O_{k,q}(x) O_{k,q}^\dagger(y), \quad (16)$$

$$h_R^m = - \sum_{x \neq y \in \Lambda_R} \sum_{k=1}^n \sum_{q=-k}^k I_k(|x-y|) O_{k,q}(x) O_{k,q}^\dagger(y), \quad (17)$$

$$h_C^{2m} = \sum_{k=1}^n (-1)^{k-1} \mathbf{O}_k^L \cdot \mathbf{O}_k^{R\dagger}, \quad (18)$$

with

$$\begin{aligned} \mathbf{O}_k^L &= (O_{k,k}^L, O_{k,k-1}^L, \dots, O_{k,-k}^L) \\ &= \sum_{x \in \Lambda_L} \alpha_k(x) (O_{k,k}(x), O_{k,k-1}(x), \dots, O_{k,-k}(x)), \end{aligned} \quad (19)$$

$$\begin{aligned} \mathbf{O}_k^R &= (O_{k,k}^R, O_{k,k-1}^R, \dots, O_{k,-k}^R) \\ &= \sum_{x' \in \Lambda_R} \alpha_k(x') (O_{k,k}(x'), O_{k,k-1}(x'), \dots, O_{k,-k}(x')). \end{aligned} \quad (20)$$

$h_{L(R)}^m$ is a collection of bonds between sites $x, y \in \Lambda_{L(R)}$ with m sites. h_C^{2m} contains parallel and crossing bonds between sites $x \in \Lambda_L$ and $y \in \Lambda_R$ which have $2m$ sites. Parallel bonds are interactions between $x \in \Lambda_L$ and $y = x' \in \Lambda_R$. Here x' means a reflection symmetric lattice point of x about the symmetry plane. So parallel bonds are perpendicular to the symmetry plane. Crossing bonds between $x \in \Lambda_L$ and $y \neq x' \in \Lambda_R$ are not perpendicular to it. $\mathbf{O}_k^{L(R)}$ has $2k+1$ components which are given by, for $-k \leq q \leq k$, $\sum_{x \in \Lambda_{L(R)}} \alpha_k(x) O_{k,q}(x)$ with real coefficients $\alpha_k(x) = \alpha_k(x')$. In Hamiltonian (3) interaction coefficients $-I_k(|x-y|)$ depend on distance between sites x and y , but, for the subsequent discussions, we consider Hamiltonian (15) containing site-dependent interactions in h_C^{2m} .

To clarify the setup of the even numbered rings, we explain examples of h_{ring}^{2m} . In the case of $m=1$ h_{ring}^2 is just one parallel bond, $m=2$ single square with two parallel bonds and two crossing bonds, and $m=3$ single hexagon with three parallel bonds and six crossing bonds. As one of examples, h_{ring}^6 is illustrated in FIG. 1. For $m \geq 4$ they are given by the same manner.

Let us perform unitary transformation on Hamiltonian

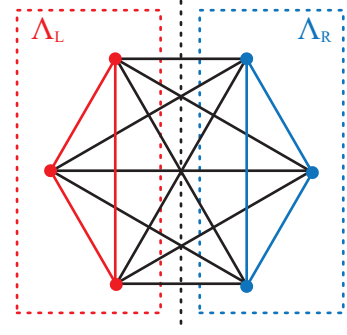


FIG. 1. Even numbered rings Λ split into equal parts Λ_L and Λ_R which are mirror images of one another with respect to symmetry plane denoted by the black dashed line. As an example of the setup of even numbered rings, h_{ring}^6 is shown. It is decomposed as h_L^3 , h_R^3 , and h_C^6 , which limitedly contains red, blue, and black bonds denoted by solid lines, respectively.

(15):

$$\begin{aligned} \tilde{h}_{\text{ring}}^{2m} &= U^\dagger h_{\text{ring}}^{2m} U \\ &= h_L^m + h_R^m - \sum_{k=1}^n \sum_{q=-k}^k O_{k,q}^L O_{k,q}^R, \end{aligned} \quad (21)$$

with

$$U = \exp \left(i\pi \sum_{x \in \Lambda_R} S_2(x) \right), \quad (22)$$

where we have used

$$U^\dagger O_{k,q}^\dagger(x) U = (-1)^k O_{k,q}(x), \quad (23)$$

for $x \in \Lambda_R$. The matrix elements of all matrices appearing in $\tilde{h}_{\text{ring}}^{2m}$ are real since $O_{k,\pm k}$ are given by the k -th power of S_\pm with real matrix elements and $O_{k,q}$ are generated by the repeated use of commutation relation (7) between $O_{k,\pm k}$ and S_\pm which have real matrix elements.

Now we can write a ground state of $\tilde{h}_{\text{ring}}^{2m}$,

$$\tilde{\psi} = \sum_{\alpha, \beta} c_{\alpha\beta} \psi_\alpha^L \otimes \psi_\beta^R, \quad (24)$$

with real coefficient matrix $c_{\alpha\beta}$, where $\{\psi_\alpha^L\}$ form a real orthonormal basis of S_3 eigenstates for the left subsystem and $\{\psi_\beta^R\}$ are the corresponding states for the right subsystem. The ground state energy of $\tilde{h}_{\text{ring}}^{2m}$ is given by

$$\begin{aligned} \langle \tilde{\psi} | \tilde{h}_{\text{ring}}^{2m} | \tilde{\psi} \rangle &= \text{Tr} c^\dagger h_L^m + \text{Tr} c^\dagger h_R^m \\ &\quad - \sum_{k=1}^n \sum_{q=-k}^k \text{Tr} c^\dagger O_{k,q}^L c O_{k,q}^{R\dagger}, \end{aligned} \quad (25)$$

with

$$(h_L^m)_{\alpha\gamma} = \langle \psi_\alpha^L | h_L^m | \psi_\gamma^L \rangle, \quad (26)$$

$$(O_{k,q}^L)_{\alpha\gamma} = \left\langle \psi_\alpha^L \left| \sum_{x \in \Lambda_L} \alpha_k(x) O_{k,q}(x) \right| \psi_\gamma^L \right\rangle, \quad (27)$$

and similarly for $(h_R^m)_{\beta\delta}$ and $(O_{k,q}^R)_{\beta\delta}$.

Following the arguments in references [28–30, 34, 35] we set $c \rightarrow c^\dagger$, then the right hand side of equation (25) is written as

$$\text{Tr} c^\dagger c h_L^m + \text{Tr} c c^\dagger h_R^m - \sum_{k=1}^n \sum_{q=-k}^k \text{Tr} c^\dagger O_{k,q}^R c O_{k,q}^{L\dagger}, \quad (28)$$

where we have used equation (5) and the cyclic property of trace in the third term. Here we note that Hamiltonian (21) is left-right symmetric. So we can see that the ground state energy is unchanged and eigenstates with coefficient matrices c^\dagger and $c + c^\dagger$ are also ground states. There exists at least one ground state with Hermite coefficient matrix. Hermite coefficient matrix can be diagonalized and then the third term in the right-hand side of equation (25) are written as

$$- \sum_{k=1}^n \sum_{q=-k}^k \sum_{l,m} c_{ll} c_{mm} |(O_{k,q})_{lm}|^2, \quad (29)$$

in the diagonal basis of c . This expression is bounded below by

$$- \sum_{k=1}^n \sum_{q=-k}^k \sum_{l,m} |c_{ll}| |c_{mm}| |(O_{k,q})_{lm}|^2. \quad (30)$$

So we can confirm that an eigenstate with positive semidefinite coefficient matrix $|c| = \sqrt{c^2}$ is a ground state of $\tilde{h}_{\text{ring}}^{2m}$.

B. Singlet Ground States

In this subsection, at first, we confirm that a ground state of h_{ring}^{2m} , i.e., $\psi = U\tilde{\psi}$ with positive semidefinite coefficient matrix $|c|$ has $S^{\text{tot}} = 0$.

Let us introduce a tensor product of spin singlet state:

$$\psi_0 = \bigotimes_{x \in \Lambda_L} \sum_{M=-S}^S (-1)^{S-M} |S, M\rangle_x \otimes |S, -M\rangle_{x'}, \quad (31)$$

where M is eigenvalues of $S_3(x)$ and $x \in \Lambda_L$ counts every pair x and its reflection symmetric point x' . Ground state $\tilde{\psi}$ with $|c|$ is written as

$$\tilde{\psi} = \sum_{\alpha} |c|_{\alpha\alpha} \psi_{\alpha}^L \otimes \psi_{\alpha}^R. \quad (32)$$

We can easily see

$$\langle \psi_0 | \psi \rangle = \langle \psi_0 | U | \tilde{\psi} \rangle = \text{Tr} |c| > 0. \quad (33)$$

Since ψ_0 has $S^{\text{tot}} = 0$ and S^{tot} is a good quantum number, ground state ψ must take $S^{\text{tot}} = 0$. Thus we can find that there exists at least one ground state with $S^{\text{tot}} = 0$. This result makes strong in the following argument.

Next, we show that all ground states of h_{ring}^{2m} have $S^{\text{tot}} = 0$ even if the ground state is degenerate. Let $b(x)$ be a real valued function of site x . Now we consider the unitarily transformed Hamiltonian under site-dependent field given by

$$\begin{aligned} \tilde{h}_{\text{ring}}^{2m}(b) &= h_L^m + h_R^m \\ &- \left[\sum_{k \neq l}^n \sum_{q=-k}^k O_{k,q}^L O_{k,q}^R + \sum_{q \neq 0} O_{l,q}^L O_{l,q}^R \right. \\ &+ \left(O_{l,0}^L - \sum_{x \in \Lambda_L} b(x) \right) \left(O_{l,0}^R - \sum_{x' \in \Lambda_R} b(x') \right) \\ &- \frac{1}{2} \left(O_{l,0}^L - \sum_{x \in \Lambda_L} b(x) \right)^2 + \frac{1}{2} (O_{l,0}^L)^2 \\ &\left. - \frac{1}{2} \left(O_{l,0}^R - \sum_{x' \in \Lambda_R} b(x') \right)^2 + \frac{1}{2} (O_{l,0}^R)^2 \right] \quad (34) \end{aligned}$$

Here we note that $\tilde{h}_{\text{ring}}^{2m}(0) = U^\dagger h_{\text{ring}}^{2m} U$. Following the argument of Kennedy-Lieb-Shastry with a trace inequality [34, 36], we get

$$e_{\text{ring}}^{2m}(b) \geq e_{\text{ring}}^{2m}(0), \quad (35)$$

concerning for the ground state energy of $\tilde{h}_{\text{ring}}^{2m}(b)$. It is required for establishment of this inequality to satisfy the conditions: the matrix elements of the matrix representations of $O_{k,q}^L$ and $O_{k,q}^R$ are real and the coefficients of all interaction terms $O_{k,q}^L O_{k,q}^R$ are negative.

When we choose

$$b(x) = \begin{cases} b & x \in \Lambda_L, \\ -b & x \in \Lambda_R \end{cases} \quad (36)$$

in equation (34), it becomes

$$\tilde{h}_{\text{ring}}^{2m}(b) = \tilde{h}_{\text{ring}}^{2m} - 2mb (O_{l,0}^L - O_{l,0}^R) + 2m^2 b^2. \quad (37)$$

Let $\tilde{\psi}(b)$ be a ground state of $\tilde{h}_{\text{ring}}^{2m}(b)$. By the variational principle and inequality (35), we have

$$\begin{aligned} \langle \tilde{\psi}(0) | \tilde{h}_{\text{ring}}^{2m}(b) | \tilde{\psi}(0) \rangle &\geq \langle \tilde{\psi}(b) | \tilde{h}_{\text{ring}}^{2m}(b) | \tilde{\psi}(b) \rangle \\ &= e_{\text{ring}}^{2m}(b) \geq e_{\text{ring}}^{2m}(0), \quad (38) \end{aligned}$$

which leads to

$$-mb \langle \tilde{\psi}(0) | O_{l,0}^L - O_{l,0}^R | \tilde{\psi}(0) \rangle + m^2 b^2 \geq 0. \quad (39)$$

This result is independent of value of b . In order to establish this inequality for arbitrary value of b it must be

$$\langle \tilde{\psi}(0) | O_{l,0}^L - O_{l,0}^R | \tilde{\psi}(0) \rangle = 0. \quad (40)$$

Noting that $U\tilde{\psi}(0) = \psi(0)$ is ψ , then we get

$$\begin{aligned} \left\langle \psi \left| \sum_{x \in \Lambda_L} \alpha_l(x) O_{l,0}(x) \right. \right. \\ \left. \left. - (-1)^l \sum_{x' \in \Lambda_R} \alpha_l(x') O_{l,0}(x') \right| \psi \right\rangle = 0. \quad (41) \end{aligned}$$

Setting $l = 1$, we can see $\langle \psi | S_3^{\text{tot}} | \psi \rangle = 0$ since the above equation must establish arbitrary values of $\alpha_1(x)$ for all x . By the rotational invariance of h_{ring}^{2m} as is shown in equation (14), this result also holds for S_1^{tot} and S_2^{tot} . So it concludes that the all ground states have $S^{\text{tot}} = 0$.

IV. CONSTRUCTIONS OF MODELS ON LATTICES WITH LOCAL HAMILTONIAN ON EVEN NUMBERED RINGS

In the previous section we have showed that all ground states of h_{ring}^{2m} on even numbered rings have $S^{\text{tot}} = 0$. In this section we consider constructions of global Hamiltonian on two dimensional lattices with local Hamiltonian h_{ring}^{2m} . Here we suppose that whole lattices are constructed with even numbered rings, such as square lattice, honeycomb lattice, 1/5-depleted lattice (CaV₄O₉).[33] 1/5-depleted lattice consists of squares and octagons.

In subsection IV A, we show that all ground states of global Hamiltonian with site-dependent interactions possesses $S^{\text{tot}} = 0$. In subsection IV B, we consider models on lattices without crossing bonds as a special case of global Hamiltonian in subsection IV A and determine conditions realizing spatially isotropic interactions. In subsection IV C and IV D, as examples of lattices with crossing bonds in this framework, we perform the same procedure in subsection IV B in the case of square and honeycomb lattices with crossing bonds.

A. Generalized lattices

In section III we have treated h_{ring}^{2m} on even numbered rings and have showed that their ground states possess $S^{\text{tot}} = 0$. It is straightforward to prove that ground states of global Hamiltonian on generalized lattices have the same result. In this subsection we shortly explain it as follows.

Let us write global Hamiltonian:

$$\mathcal{H} = \mathcal{H}_L + \mathcal{H}_R + \mathcal{H}_C. \quad (42)$$

\mathcal{H}_C is constructed with translated copies of local Hamiltonian $\sum_j^m h_C^{2j}$ on an even numbered ring with $2m$ sites in the direction parallel to the symmetry plane,[37] i.e.,

$$\mathcal{H}_C = \sum_{\Lambda_C} \sum_j^m h_C^{2j}, \quad (43)$$

where the summation for j is taken if it is necessary and the same applies hereinafter. The sites belonging to Λ_C are denoted by the collection of sites which are translated copies of sites belonging to an even numbered ring such as single square and single hexagon in the direction parallel to the symmetry plane(see FIG.2).

Let $h_{\text{ring}}^{2m} = h_L^m + h_R^m + \sum_j^m h_C^{2j}$. $\mathcal{H}_{L(R)}$ consists of the collection of translated copies of h_{ring}^{2m} on $\Lambda_{L(R)}$ and the

collection of bonds in $h_{L(R)}^m$ on $\partial\Lambda_{L(R)}$. The sites belonging to $\partial\Lambda_{L(R)}$ is denoted by $\Lambda_{L(R)} \cap \Lambda_C$ (the sites belonging to $\Lambda_C = \partial\Lambda_L \cup \partial\Lambda_R$). Then, global Hamiltonian on $\Lambda_{L(R)}$ is written as

$$\mathcal{H}_{L(R)} = \sum_{\Lambda_{L(R)}} h_{\text{ring}}^{2m} + \sum_{\partial\Lambda_{L(R)}} h_{L(R)}^m. \quad (44)$$

The second term in this Hamiltonian is omitted if global Hamiltonian is constructed with bond sharing even numbered rings. These operations should be realized to construct the global Hamiltonian on the two dimensional lattices $\Lambda = \Lambda_L \cup \Lambda_R$. We recall that Λ_L and Λ_R are equal parts and Λ has reflection symmetry about the symmetry plane. Here we use ‘generalized’ lattices in the sense that global Hamiltonian (42) is constructed with h_C^{2j} containing site-dependent interactions in spite that \mathcal{H}_n has spatially isotropic interactions.

We can easily see that global Hamiltonian (42) also holds the same results in section III. Roughly speaking, main differences are that, in equations (34) and (35), $h_{L(R)}^m$ and local ground state ψ are replaced by $\mathcal{H}_{L(R)}$ and global ground state $\Psi = \sum_{\alpha,\beta} C_{\alpha\beta} \Psi_\alpha^L \otimes \Psi_\beta^R$ and $\sum_{\Lambda_C} \sum_j^m$ appears in the terms of parallel and crossing bonds on Λ_C . Through the same procedure in subsection III B we can see similar equation with respect to $\Lambda_C (= \partial\Lambda_L \cup \partial\Lambda_R)$ to equation (41) as follows.[48]

$$\left\langle \Psi \left| \sum_{x \in \partial\Lambda_L} \alpha(x) O_{l,0}(x) - (-1)^l \sum_{x' \in \partial\Lambda_R} \alpha(x') O_{l,0}(x') \right| \Psi \right\rangle = 0 \quad (45)$$

and

$$\left\langle \Psi \left| \sum_{x \in \partial\Lambda_L} O_{l,0}(x) - (-1)^l \sum_{x' \in \partial\Lambda_R} O_{l,0}(x') \right| \Psi \right\rangle = 0. \quad (46)$$

Let us set $l = 1$ and impose a periodic boundary condition in the direction perpendicular to the symmetry plane, we conclude that all ground states of global Hamiltonian have $S^{\text{tot}} = 0$ if whole lattices can be constructed with translated copies of Λ_C . Otherwise, we need to set other symmetry planes and impose periodic boundary conditions in the directions perpendicular to those symmetry planes.

B. Lattices without crossing bonds

In the case of bipartite lattices, whole lattices are constructed with even numbered rings without crossing bonds. so we can construct global Hamiltonian with $h_{\text{ring}}^2(m = 1)$, i.e., nearest neighbor pairs $\langle x, y \rangle$ only. This

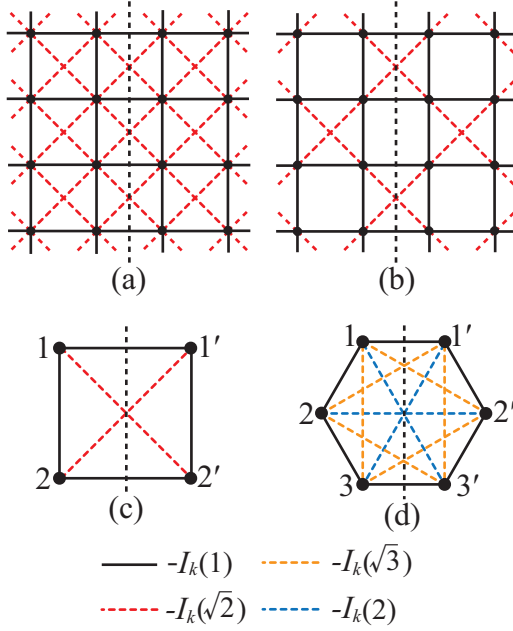


FIG. 2. (a) Square lattice with crossing bonds. (b) Checkerboard type. (c) Local Hamiltonian (48) on single square with crossing bonds. Black solid and red dashed lines represent nearest neighbor interaction $-I_k(1)$ and second neighbor interaction $-I_k(\sqrt{2})$, respectively. (d) Local Hamiltonian (55) on single hexagon with crossing bonds. Black solid, orange dashed, and blue dashed lines represent nearest neighbor interaction $-I_k(1)$, second neighbor interaction $-I_k(\sqrt{3})$, and third neighbor interaction $-I_k(2)$, respectively. Black dashed lines in FIGs (a)-(d) represent reflection symmetric plane.

simplest model of Hamiltonian (42) is written as

$$\begin{aligned} & \sum_{\langle x,y \rangle \in \Lambda} h_{\text{ring}}^2 \\ &= \sum_{\langle x,y \rangle \in \Lambda} \sum_{k=1}^n (-1)^{k-1} \alpha_k(x)^2 \sum_{q=-k}^k O_{k,q}(x) O_{k,q}^\dagger(y). \end{aligned} \quad (47)$$

Interaction coefficients in the right hand side of this equation are correspond to $-I_k(1)$ and we see $(-1)^k I_k(1) \geq 0$ for each k .

C. Square lattices with crossing bonds

In references [28, 29, 38, 39] the antiferromagnetic Heisenberg models with next nearest neighbor interactions on the square lattice and its checkerboard type were discussed. In this subsection, following these previous studies we treat the case of Hamiltonian (3).

Now let us consider constructions of global Hamiltonian on the square lattice with $h_{\text{ring}}^4 (m=2)$. h_{ring}^4 is local Hamiltonian on single square with crossing bonds (see FIG.3-(c)). To determine the condition realizing spa-

tially isotropic interactions of the models we write

$$\begin{aligned} & h_{\text{ring}}^4 - \sum_{k=1}^n \sum_{q=-k}^k (-1)^{k-1} \sum_{x=1}^2 \alpha_k(x)^2 O_{k,q}(x) O_{k,q}^\dagger(x') \\ & - \sum_{k=1}^n \sum_{q=-k}^k I_k(1) \sum_{x=1}^2 O_{k,q}(x) O_{k,q}^\dagger(x') \\ & = h_{\text{ring}}^4 + \sum_{k=1}^n \sum_{q=-k}^k \sum_{x=1}^2 [-I_k(1) \\ & - (-1)^{k-1} \alpha_k(x)^2] O_{k,q}(x) O_{k,q}^\dagger(x'), \end{aligned} \quad (48)$$

where $-I_k(1)$ are denoted by coefficients of the first neighbor 2^k -pole interactions. To explain the lattice structures and their spatial isotropy, they are illustrated in FIGs 2-(a),(b),(c). Noting that the factor $(-1)^k$ appears from each term in the right hand side of this equation through the unitary transformation, then it can be seen as a combination of h_{ring}^4 and h_{ring}^2 if interaction coefficients of these local Hamiltonian satisfy

$$-(-1)^k I_k(1) + \alpha_k(1)^2 \leq 0, \quad (49)$$

$$-(-1)^k I_k(1) + \alpha_k(2)^2 \leq 0, \quad (50)$$

for each k and we get

$$(-1)^k I_k(1) \geq 0, \quad (51)$$

$$-I_k(1) \leq -I_k(\sqrt{2}) \leq I_k(1), \quad (52)$$

with second neighbor interaction coefficients $-I_k(\sqrt{2}) = (-1)^{k-1} \alpha_k(1) \alpha_k(2)$.

When global Hamiltonian is constructed with local Hamiltonian (48) on bond sharing squares as in FIG 2-(a), the results in subsection IV A hold if $-I_k(1)/2 \leq I_k(\sqrt{2}) \leq I_k(1)/2$ and $(-1)^k I_k(1) \geq 0$. But its checkerboard type is site sharing as in FIG 2-(b). So condition $-I_k(1) \leq I_k(\sqrt{2})/2 \leq I_k(1)$ is replaced by $-I_k(1) \leq I_k(\sqrt{2}) \leq I_k(1)$.

D. Honeycomb lattice with crossing bonds

In this subsection we treat models on the honeycomb lattice with crossing bonds which is constructed with translated copies of local Hamiltonian on single hexagon (FIG 2-(d)). Let us consider global Hamiltonian with \mathcal{H}_C given by

$$\sum_{\Lambda_C} \sum_{j=2}^3 h_C^{2j} = \sum_{\Lambda_C} (h_C^6 + h_C^4). \quad (53)$$

In FIG. 2-(d) h_C^4 is local Hamiltonian on a rectangular formed by four sites 1, 1', 3, 3' except dashed orange bonds. So inner product of \mathcal{O}_k^L and $\mathcal{O}_k^{R\dagger}$ in h_C^4 is defined by

$$\mathcal{O}_k^L = \sum_{x=1,3} \beta_k(x) (O_{k,k}(x), O_{k,k-1}(x), \dots, O_{k,-k}(x)) \quad (54)$$

and O_k^R with $\beta_k(x')$ on sites $1', 3'$.

Following the previous subsection let us write the local Hamiltonian:

$$\begin{aligned}
& h_{\text{ring}}^6 - \sum_{k=1}^n \sum_{q=-k}^k (-1)^{k-1} \sum_{x=1}^3 \alpha_k(x)^2 O_{k,q}(x) O_{k,q}^\dagger(x') \\
& - \sum_{k=1}^n \sum_{q=-k}^k \sum_{x=1}^3 I_k(|x-x'|) O_{k,q}(x) O_{k,q}^\dagger(x') \\
& + h_C^4 - \sum_{k=1}^n \sum_{q=-k}^k (-1)^{k-1} \sum_{x=1,3} \beta_k(x)^2 O_{k,q}(x) O_{k,q}^\dagger(x') \\
& = h_{\text{ring}}^6 + h_C^4 \\
& + \sum_{k=1}^n \sum_{q=-k}^k \left\{ [-I_k(2) - (-1)^{k-1} \alpha_k(2)^2] O_{k,q}(2) O_{k,q}^\dagger(2') \right. \\
& + \sum_{x=1,3} [-I_k(1) - (-1)^{k-1} \alpha_k(x)^2 \\
& \left. - (-1)^{k-1} \beta_k(x)^2] O_{k,q}(x) O_{k,q}^\dagger(x') \right\}, \quad (55)
\end{aligned}$$

where $-I_k(1)$, $-I_k(\sqrt{3})$, and $-I_k(2)$ are denoted by first, second, and third neighbor interaction coefficients of 2^k -pole interactions, respectively, as illustrated in FIG. 2-(d). Similar to the previous subsection, right hand side of this equation can be seen as a combination of h_{ring}^6 and h_C^4 along with h_{ring}^2 and its analogue if

$$-(-1)^k I_k(2) + \alpha_k(2)^2 \leq 0, \quad (56)$$

$$-(-1)^k I_k(1) + \alpha_k(1)^2 + \beta_k(1)^2 \leq 0, \quad (57)$$

$$-(-1)^k I_k(1) + \alpha_k(3)^2 + \beta_k(3)^2 \leq 0, \quad (58)$$

and conditions on interaction parameters satisfying spatial isotropy in Λ are given by

$$-I_k(\sqrt{3}) = (-1)^{k-1} \alpha_k(1) \alpha_k(2) = (-1)^{k-1} \alpha_k(2) \alpha_k(3), \quad (59)$$

$$-I_k(2) = (-1)^{k-1} [\alpha_k(1) \alpha_k(3) + \beta_k(1) \beta_k(3)], \quad (60)$$

$\alpha_k(1) = \alpha_k(3)$, and $\beta_k(1) = \beta_k(3)$ for each k . From these equations and inequalities we get

$$\frac{1}{2} \geq \frac{I_k(2)}{I_k(1)} \geq -\frac{1}{4} + \sqrt{\frac{1}{16} + 2 \left(\frac{I_k(\sqrt{3})}{I_k(1)} \right)^2}, \quad (61)$$

$$(-1)^k \frac{I_k(1)}{2} \geq (-1)^k I_k(2) \geq 0, \quad (62)$$

where we have replaced $I_k(1)$ by $I_k(1)/2$ since global Hamiltonian is constructed with local Hamiltonian (55) on bond sharing hexagons.

V. SUMMARY AND DISCUSSIONS

We have discussed the Heisenberg models with higher order interactions or multipole interactions on finite lattices with reflection symmetry written as in the form of

Hamiltonian (15) or (42) and have found that there exists at least one ground state with $S^{\text{tot}} = 0$. Moreover imposing a periodic boundary condition in the direction perpendicular to the symmetry plane, we have confirmed that the all ground states possess $S^{\text{tot}} = 0$ even if the ground state is degenerate. These results are a straightforward extension of the Lieb-Schupp theorem to these models.

For establishment of the results in subsections III B and IV A we did not put any restrictions on signs or values of interaction coefficients $-I_k(|x-y|)$ of h_L^m and h_R^m except their reflection symmetry (see Hamiltonian (15)-(18) and (42)-(44)). These coefficients are not essential to our results. In subsections IV B, IV C, and IV D they were determined by the conditions in order that \mathcal{H}_n possesses spatially isotropic interactions as in FIG. 2. On the other hand restrictions on interaction coefficients of parallel bonds and crossing bonds in h_C^{2m} come from the establishment of inequality (35) and similar inequalities for the models in this paper.[48] Therefore we have no idea for improvement of these restrictions.

In this section, we summarize and discuss the results in section III and IV which are divided into the models on lattices with and without crossing bonds.

A. Lattices without crossing bonds

In this case, models are constructed with local Hamiltonian h_{ring}^2 . Typical examples of the whole lattice are bipartite lattices such as hypercubic lattice, honeycomb lattice, and 1/5-depleted lattice. In the case of the 1/5-depleted lattice we should set the symmetry plane which intersects octagons and squares. The results hold if $(-1)^k I_k(1) \geq 0$ for each k .

In the following we shall explain comparisons with the results of the Marshall-Lieb-Mattis type argument. $H_1 = \mathcal{H}_1$ with $-J_1(1) = -I_1(1) > 0$ is the spin- S antiferromagnetic Heisenberg model. The Marshall-Lieb-Mattis theorem assures that its ground state is unique and has $S^{\text{tot}} = 0$. So the result given by the Lieb-Schupp theorem is completely covered by the Marshall-Lieb-Mattis theorem with uniqueness of the ground state.

H_2 is equivalent to bilinear $-J_1(1)(= -I_1(1) - 3I_2(1)/4)$ and biquadratic $-J_2(1)(= -3I_2(1)/2)$ exchange interaction model H_2 . From the previous studies of the Marshall-Lieb-Mattis type argument it was known that the same results hold for H_2 with $S = 1$ in the region $J_2(1) > J_1(1)$, $J_2(1) \geq 0 (J_1(1) \neq 0)$ and with $S > 1$ in the region $0 \leq J_2(1) \leq -J_1(1)/2S(S-1)$. [23-25] On the other hand, our results based on the Lieb-Schupp theorem show that all ground states have $S^{\text{tot}} = 0$ in the region $J_1(1) \geq 2J_2(1)$, $J_2(1) \geq 0$ for any S . For $S = 1$ purely biquadratic interaction model ($J_1(1) = 0$, $J_2(1) > 0$) satisfies SU(3) symmetry and its ground state is degenerate. Our results can conclude that all degenerate ground states possess $S^{\text{tot}} = 0$. In the case of $S > 1$ our results extend the region which one can conclude ground

states with $S^{\text{tot}} = 0$. These results are summarized in FIG. 3.

For \mathcal{H}_n with $n > 2$, our results also hold if $(-1)^k I_k(1) \geq 0$ for each k . As far as we know, the results of the Marshall-Lieb-Mattis type argument does not exist.

Our study is concerned with models on finite lattices with reflection symmetry and their ground states possess $S^{\text{tot}} = 0$, but in infinite volume limit continuous symmetry breaking may occur. The antiferromagnetic Heisenberg model on bipartite lattices in two or more dimensions is known to exhibit Néel long range order in its ground state. For the d -dimensional hypercubic lattice, it was rigorously proved in $d \geq 3$ for any S and in $d = 2$ for $S \geq 1$ [34, 40, 41], and for the honeycomb lattice for $S \geq 3/2$ [1]. Ground state phase diagram of $S = 1$ bilinear-biquadratic model (H_2) with $J_2(1) > 0$ on the square or the simple cubic lattice is considered as follows. [42–45] The region $J_2(1) > J_1(1) > 0$ is the ferroquadrupole (spin nematic) phase, $J_1(1) < 0$ the Néel ordered phase, and $0 < J_2(1) < J_1(1)$ the ferromagnetic phase, which are illustrated in FIG. 3-(a). In our parameter region there exist Néel long range order and ferroquadrupole long range order in infinite volume ground state, which were rigorously proved in parts of these parameter region. [36, 46, 47] These proofs were given by the method of infrared bounds whose key inequality is upper bound on the Fourier transformed correlation function in whole momentum space which is derived from inequality (35) and similar ones for the ground state energy or analogous inequality for the partition function.

B. Lattices with crossing bonds

In this case, models are constructed with translated copies of h_{ring}^{2m} with $m > 1$ on rings. These rings have crossing bonds. In subsection IV A we have proved that global Hamiltonian (42) holds the same results in subsection III B. These models have site-dependent interactions. So in subsections IV C and IV D we considered conditions which Hamiltonian (42) possesses spatially isotropic interactions on the square and the honeycomb lattices with crossing bonds as in FIG. 2. For the non-checkerboard square lattice it is realized in the region $-I_k(1)/2 \leq I_k(\sqrt{2}) \leq I_k(1)/2$ and checkerboard type $-I_k(1) \leq I_k(\sqrt{2}) \leq I_k(1)$ for each k . The result for the honeycomb lattice is given by inequalities (61) (62), and setting $|I_k(1)| = 1$ it is illustrated in FIG. 4.

Only about the frustrated antiferromagnetic Heisenberg model on the square and honeycomb lattice with crossing bonds, we explain relation between our results on finite lattices and the results of various theoretical studies on infinite lattices. There exist detailed reviews of these models on square lattices in the paper [49] (see also references therein). In infinite volume limit, within our parameter region, the ground state phase diagram expected to be valid is summa-

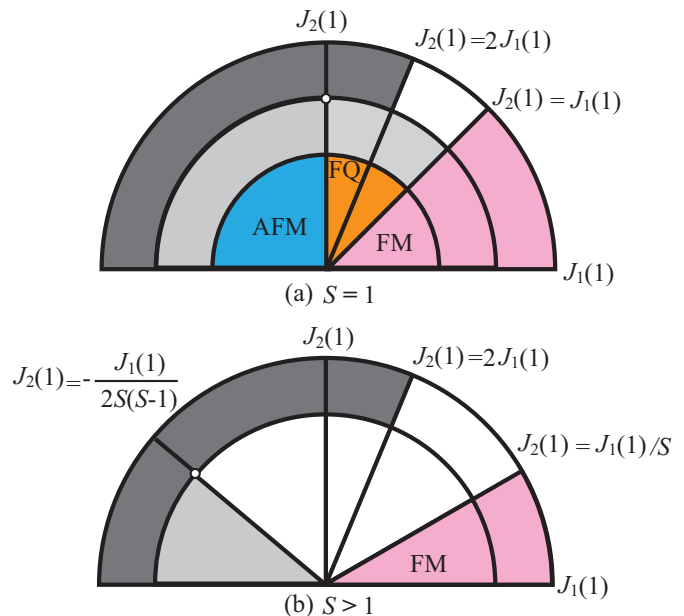


FIG. 3. (a) $S = 1$ and (b) $S > 1$ ground state phase diagram of the bilinear-biquadratic exchange Hamiltonian on finite lattices without crossing bonds. Our results based on the Lieb-Schupp theorem mean that all ground states have $S^{\text{tot}} = 0$ in the dark gray region. The results of the Marshall-Lieb-Mattis type arguments mean that unique ground state has $S^{\text{tot}} = 0$ in the light gray region. Three colored regions in inner semicircle in FIG. (a) are expected phase diagram of $d \geq 2$ dimensional hyper cubic lattice in infinite volume limit. Pink, orange, and blue region is ferromagnetic (FM), ferroquadrupole (FQ), and Néel ordered (AFM) phase, respectively.

rized as follows. For the non-checkerboard (checkerboard) square lattice, in the case of $S = 1/2$, the region $0(0) \leq J_1(\sqrt{2})/J_1(1) \lesssim 0.4(0.8)$ is the Néel ordered phase and $0.4(0.8) \lesssim J_1(\sqrt{2})/J_1(1) \leq 0.5(1)$ the quantum paramagnetic phase without magnetic long range order, and in $S = 1$, $0(0) \leq J_1(\sqrt{2})/J_1(1) \leq 0.5(1)$ the Néel ordered phase. In the case of $S = 1$ and non-checkerboard type, by using the method of infrared bounds, the existence of Néel long range order was rigorously proved in small $J_1(\sqrt{2})/J_1(1)$. [38] Ground state phase diagram of $S = 1/2$ on the honeycomb lattice (non-checkerboard type) was obtained in reference [50, 51]. Our parameter region also seems to be contained in the Néel ordered phase and the quantum paramagnetic phase.

In this paper we focus on one dimensional rings and two dimensional lattices. Application of these results to three dimensional lattices can be easily extended. In that case, as a simplest example, we can consider local Hamiltonian on single cubes with crossing bonds and it should be written as h_{box}^8 .

Lieb-Schupp called FIG. 2-(b) pyrochlore checkerboard since it is a two dimensional projection of a three dimensional pyrochlore lattice. But this framework is not ap-

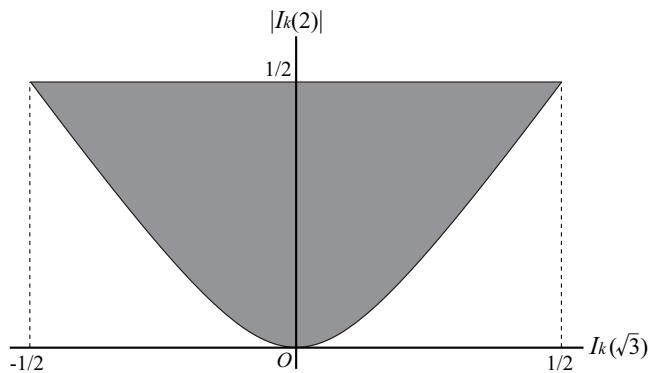


FIG. 4. Ground states of \mathcal{H}_n on honeycomb lattice with crossing bonds have $S^{\text{tot}} = 0$ in the gray parameter region for each k which is determined by inequalities (61) and (62). $I_k(1)$ is scaled as -1 if k is odd, otherwise $I_k(1) = 1$, and $-1/2 \leq I_k(2) \leq 0$ if k is odd, otherwise $0 \leq I_k(2) \leq 1/2$.

plicable to the pyrochlore lattice unfortunately, because it lacks reflection symmetry. They also called equation (41) with $l = 1$ quantum analogue of ice rule in the context of the correspondence between Ising like ferromagnet with crystal field anisotropy on the pyrochlore lattice and configuration of four hydrogen atoms around an oxygen atom in ice.[28, 29] In that point of view equations (41) and (46) are generalization of ice rule to any even numbered frustrated units and 2^{2l-1} pole moment higher than dipole.

In the following we shall comment on effects of crossing bonds on stability of the Néel ordered phase and the ferroquadrupole (spin nematic) phase. As was illustrated in FIG. 3-(a), there exist the Néel ordered phase and the ferroquadrupole phase which are separated by the line at $J_1(1) = 0$. By adding antiferromagnetic crossing bonds to the square lattice, it is clear that the Néel order exhibiting anti-alignment of spin is not stable. On the other hand, the quadrupole order is not alignment of spin but nematicity of spin, and from equation (23) it can be seen that $O_{k,0}$ with even k is even parity with respect to time reversal. The ferroquadrupole order is uniformly aligned nematic and does not seem to be suffer from geometrical frustration. So stability of the ferroquadrupole phase is not affected by frustration due to antiferromagnetic crossing bonds unlike the Néel ordered phase. Now we set next nearest neighbor inter-

actions $-J_1(\sqrt{2}) = -\alpha J_1(1)$ and $-J_2(\sqrt{2}) = -\alpha J_2(1)$ with $0 \leq \alpha \leq 1/2$, then FIG. 3-(a) is expected to be changed as follows. Phase boundary $J_2(1) = J_1(1)$ is unchanged by adding crossing bonds with ferromagnetic and ferroquadrupole interactions since the region $0 \leq J_2(1) < J_1(1)$ is saturated ferromagnetic ground state. On the other hand, phase boundary $J_1(1) = 0$ closes to the antiferromagnetic Heisenberg point as α approaches $1/2$, i.e., by adding crossing bonds with antiferromagnetic and ferroquadrupole interactions, the ferroquadrupole phase becomes dominant and the Néel ordered phase is suppressed if there do not exist different phases between these two phases. As for stability of the Néel order, the triangular and pyrochlore lattices are slightly different situation from the square lattice with antiferromagnetic crossing bonds. The ground state phase diagrams of $S = 1$ bilinear-biquadratic exchange model on the triangular and pyrochlore lattices are obtained in reference [5, 6, 16] and the same situation in the above scenario is shown.

Finally we shall propose the physical realization of the ferroquadrupole phase in magnetic materials. Usually biquadratic interaction is small as compared with bilinear interaction and the ferroquadrupole phase is unphysical in magnetic materials. In reference [3] Mila and Zhang proposed a mechanism leading to a significant biquadratic interaction in $S = 1$ systems as follows. The super exchange interaction between atoms with three orbitals and two outer electrons per atom, which consists of the two singly occupied doubly degenerate orbitals with the lowest energy and an unoccupied orbital with slightly higher energy. The virtual electron transition via the higher energy orbital favors ferromagnetic spin interaction, which compensates largely the antiferromagnetic superexchange interaction. As a result, the biquadratic interaction becomes predominant relatively. Thus we expect that highly frustrated antiferromagnetic materials with biquadratic exchange interactions originated from the Mila-Zhang mechanism may exhibit the ferroquadrupole phase.

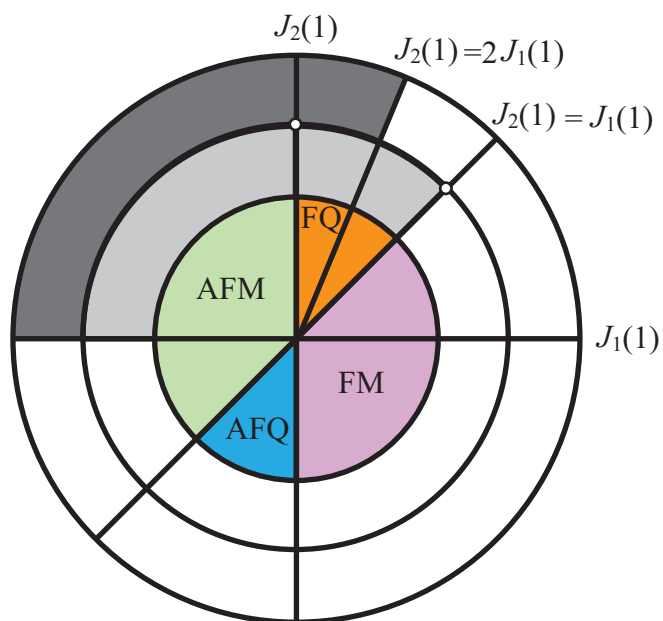
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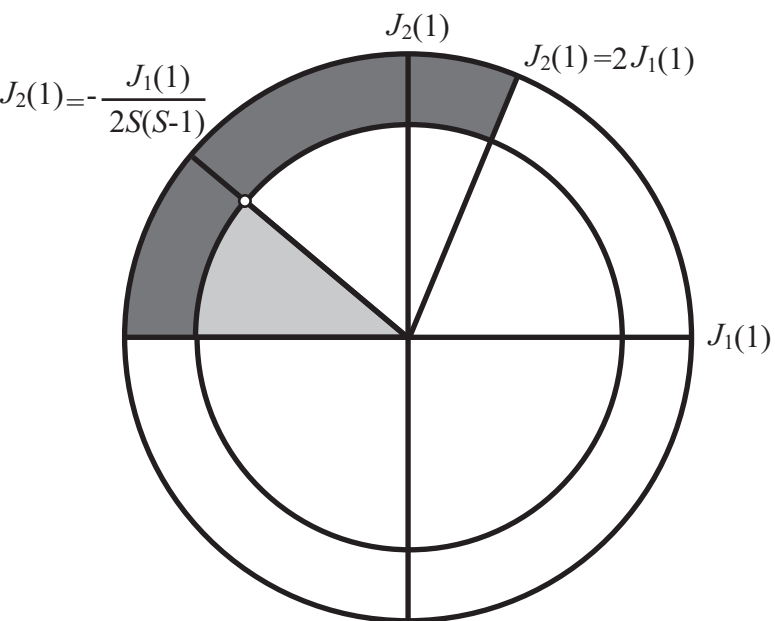
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(a) $S = 1$



(b) $S > 1$